Continuum theory of vacancy-mediated diffusion

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Abstract

We present and solve a continuum theory of vacancy-mediated diffusion (as evidenced, for example, in the vacancy driven motion of tracers in crystals). Results are obtained for all spatial dimensions, and reveal the strongly non-gaussian nature of the tracer fluctuations. In integer dimensions, our results are in complete agreement with those from previous exact lattice calculations. We also extend our model to describe the vacancy-driven fluctuations of a slaved flux line.

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I. INTRODUCTION

Random walks and associated diffusion processes are ubiquitous in Nature [1]. Our understanding of their properties has grown steadily over the years, with contributions from workers in many fields, but there are always surprises in store as we learn to ask new and more probing questions.

An interesting example of a non-trivial random walk problem is that of vacancy-mediated diffusion (VMD) [2,3]. This applies to the case of a crystal containing a low density of vacancies; the question being, what are the fluctuations of a tagged particle (or impurity) which moves only due to exchange with wandering vacancies? This process has a more generic application as it is one of the simplest 'slaved diffusion processes'. It has been studied in various guises over the years [3] and a lattice formulation was solved exactly by Brummelhuis and Hilhorst [4] in the late Eighties. The most revealing aspect of their solution is the strongly *non-gaussian* nature of the tagged particle's fluctuations. The same lattice model received alternative exact treatments recently [5].

Given one is generally interested in the scaling behaviour of such systems, it is useful to have a more coarse-grained treatment to hand which allows one to access the (hopefully universal) long-wavelength behaviour directly, without having to resort to a sophisticated lattice calculation. A well-known example of such a situation is the critical behaviour of a ferromagnet [6], which may be studied within the context of the lattice (Ising) model (where an exact treatment is only possible in dimensions $d = 1, 2$), or at a more coarse-grained level via the ϕ^4 field theory (where controlled calculations can estimate the exponents in the most interesting case $d = 3$) [7].

The purpose of this paper is to construct and analyse a continuum (or coarse-grained) theory of vacancy-mediated diffusion. Fortunately the continuum theory is exactly solvable in all dimensions, which allows a direct comparison with the previous lattice calculations [4] – complete agreement is found. Given this situation, one has confidence in applying the continuum theory to situations where a lattice formulation would be more difficult, if not

intractable. We consider one such situation here; namely the motion of a directed (flux) line under the action of diffusing vacancies.

The outline of the paper is as follows. In the next section we define the lattice model of vacancy-mediated diffusion more precisely, and, using this as a base, construct the continuum theory. We then highlight the 'mesoscopic' physics contained within this coarse-grained model. In Section III we analyse the continuum model at the level of mean-field theory (MFT). The apparent physics within MFT is simple, and the solutions of the mean-field equation yield results which contain some correct scaling information (i.e. the length-time scaling), but completely miss the more interesting statistical aspects; namely the strongly non-gaussian nature of the fluctuations as found from the lattice calculations. Thus we are led to attempt an exact solution of the continuum theory. This is made possible by formulating an infinite-order perturbation theory, as described in Section IV. The results for various dimensions are derived and presented in Section V – the non-gaussian statistics are seen to be completely reproduced, with the added advantage that the results from the continuum theory are valid for arbitrary dimension d. In Section VI we give an illustration of the utility of this coarse-grained approach. We formulate a continuum model for the transverse fluctuations of a $d + 1$ directed (flux) line due to VMD of pinning centres. We end the paper with Section VII which contains our conclusions, along with some suggested extensions of the present work.

II. FORMULATION OF THE CONTINUUM THEORY

We first consider a simple lattice formulation of VMD with which we shall motivate our continuum theory. To be specific, let us consider the following lattice model. Each site $\mathbf{r}$ of a d-dimensional hypercubic lattice contains a spin $S_{\mathbf{r}}$ which may take the values ± 1. [In the crystal lattice application one would take all spins as 'up', thus referring to crystal atoms, except for the tagged particle (or impurity) which would be assigned a 'down' spin.] A spin may only alter its value when involved in an exchange with a single diffusing vacancy

located at the position $\mathbf{R}(t)$. As only the *position* of the vacancy is relevant we may give the vacancy a spin $+1$ for notational convenience. [We shall assume for simplicity that only one vacancy is present – the generalization to many vacancies is straightforward.] There are various ways to describe this model. For instance, one can define a probability distribution for the vacancy position and the position of the 'up' and 'down' spins, and then write a master equation for its evolution. Alternatively, one can regard the process as a stochastic cellular automata (SCA), and attempt to write explicit SCA rules for its operation. We shall follow the latter approach.

The rule for updating the vacancy position is written as

$$\mathbf{R}(t+\delta t) = \mathbf{R}(t) + \mathbf{l}(t) \ , \tag{1}$$

where $\mathbf{l}(t)$ is a unit lattice vector drawn with equal probability from the $2d$ possible choices. The update of the spins is written down as follows: a spin will remain unchanged unless it is located either at the vacancy position at time t, or at the subsequent vacancy position at time $t+\delta t$. Thus we have

$$S_{\mathbf{r}}(t+\delta t) = S_{\mathbf{r}}(t) + \delta_{\mathbf{r},\mathbf{R}(t)}[S_{\mathbf{R}(t+\delta t)}(t) - S_{\mathbf{r}}(t)] + \delta_{\mathbf{r},\mathbf{R}(t+\delta t)}[S_{\mathbf{R}(t)}(t) - S_{\mathbf{r}}(t)] \ , \tag{2}$$

which may be re-arranged (with the help of (1)) in the appealing form

$$S_{\mathbf{r}}(t+\delta t) - S_{\mathbf{r}}(t) = [S_{\mathbf{R}(t)+\mathbf{l}(t)}(t) - S_{\mathbf{R}(t)}(t)]\ [\delta_{\mathbf{r},\mathbf{R}(t)} - \delta_{\mathbf{r},\mathbf{R}(t)+\mathbf{l}(t)}] \ . \tag{3}$$

The factorization of the SCA update for $S_{\mathbf{r}}(t)$ gives us reason to hope that a simple Taylor expansion may be invoked to yield an accurate continuum limit, since the leading order terms will appear in a multiplicative fashion (as opposed to terms occurring in an additive fashion, in which case one is unsure of their relative importance). Thus we shall take the simplest continuum limit: First, the position of the vacancy is described by a real vector $\mathbf{R}(t) \in \mathcal{R}^d$ satisfying

$$d\mathbf{R}/dt = \xi(t) \ , \tag{4}$$

which is the continuum equivalent of Eq.(1). The random lattice vector $\mathbf{l}$ has given way to a random vector ξ (with an implicit factor of $\sqrt{\delta t}$) which is drawn from a gaussian distribution $P[\xi]$ with zero mean, and covariance $\langle \xi^\alpha(t)\xi^\beta(t')\rangle = D\delta_{\alpha,\beta}\delta(t-t')$. [Henceforth, angle brackets denote an average over P.] Second, the spin variable $S_{\mathbf{r}}(t)$ will be replaced by a coarse-grained 'magnetization density', or 'order parameter' (OP) denoted by the field $\phi(\mathbf{r},t)$. Expanding each term of Eq.(3) to first order, we obtain

$$\partial_t \phi(\mathbf{r},t) = -\lambda \left[\xi \cdot \frac{\partial \phi(\mathbf{R}(t),t)}{\partial \mathbf{R}(t)} \right] \partial_t \delta^d(\mathbf{r}-\mathbf{R}(t)) \ , \tag{5}$$

where λ is a phenomenological parameter with dimensions $L^d T$. This last equation represents our continuum theory of VMD. It *loosely* resembles a Langevin equation, but the extremely implicit appearances of the noise ξ throughout the equation forbid such a simple designation. For instance, it is not clear how one would write down a dynamical equation for the OP probability distribution $P[\phi,t]$ – it would certainly not fit within the standard Fokker-Planck category. However, taxonomy is of little importance to us. We shall accept the equation on its own merits. As a first step in this direction, let us probe its more obvious physical content.

We can ascribe an independent meaning to each of the two factors on the right-hand-side of Eq.(5). The second of the two is simple enough in spirit – it allows temporal change of the OP only in the neighbourhood of the vacancy position $\mathbf{R}(\mathbf{t})$. The first factor guarantees (via the directional derivative) that the OP temporally changes only when the vacancy moves through a region in which the OP has spatial variation. Furthermore, the amount of temporal variation is linearly coupled to the amount of spatial variation (with a strength λ). This appears reasonable with regard to the physical properties of VMD we wish to model. Whether one would have written down such a continuum model *a priori* based on these considerations is unclear. However, given the simple 'derivation' from the SCA, one finds the *a posteriori* physical motivations satisfactory.

It is useful to note that analytic progress on Eq.(5) is very difficult without first recasting it in Fourier space (so as to rid ourselves of the implicit nature of the noise). Denoting the

Fourier transform (FT) of the OP by $\tilde{\phi}(\mathbf{k}, t)$ we have

$$\partial_t \tilde{\phi}(\mathbf{k}, t) = -\lambda \, \tilde{G}(\mathbf{k}, t) \int dk_1 \, \tilde{G}(\mathbf{k}_1, t)^* \tilde{\phi}(\mathbf{k}_1, t) \ , \tag{6}$$

where

$$\tilde{G}(\mathbf{k}, t) = (\xi \cdot \mathbf{k}) \exp[i\mathbf{k} \cdot \mathbf{R}(t)] \ , \tag{7}$$

and $dk \equiv d^d k/(2\pi)^d$.

We shall now discuss the initial conditions. As for the vacancy, we simply need to define its initial position $\mathbf{R}_0 \equiv \mathbf{R}(t = 0)$. The initial condition $\phi_0(\mathbf{r})$ for the OP will vary according to the physical system we are interested in modelling. As regards the case of a tagged particle (or impurity) being driven by the wandering vacancy, we take the initial condition on the lattice to be all spins up, bar one down spin (representing the tag) located at some point (the origin, say). In the continuum, this may be described by

$$\phi_0(\mathbf{r}) = A - B\delta^d(\mathbf{r}) \ . \tag{8}$$

Alternative scenarios may be investigated by modifying the initial OP distribution. For instance, setting ϕ_0 to be a step function would be appropriate for modelling the vacancy mediated roughening of an initially straight domain wall [8].

Once the initial conditions are defined we may formally integrate the equations of motion to give

$$\mathbf{R}(t) = \mathbf{R}_0 + \int_0^t dt' \ \xi(t') \tag{9}$$

and

$$\tilde{\phi}(\mathbf{k}, t) = \tilde{\phi}_0(\mathbf{k}) - \lambda \int_0^t dt' \ \tilde{G}(\mathbf{k}, t') \int dk_1 \ \tilde{G}(\mathbf{k}_1, t')^* \tilde{\phi}(\mathbf{k}_1, t') \ . \tag{10}$$

This completes our formulation of the model. The main focus of this paper is to calculate the mean OP density $\rho(\mathbf{r}, t) = \langle \phi(\mathbf{r}, t) \rangle$, which is the quantity analogous to the tagged particle distribution function calculated in previous lattice studies. In the next section, we shall present a brief mean-field analysis of ρ, whilst sections IV and V contain a description of the exact solution for ρ from Eqs.(9) and (10) above.

III. MEAN FIELD THEORY

The purpose of this section is to indicate how much one can learn about the system from a simple, yet uncontrolled, mean field theory (MFT). We shall find that MFT predicts the correct length-time scaling, but misses the non-gaussian nature of the fluctuations in VMD. This error persists in all dimensions (bar $d = 0$).

We shall define the MFT to be used here in an operational sense. Namely, we perform the simplest possible average of the OP equation of motion (6), by replacing the average of the right-hand-side by the product of two separate averages. Explicitly, we have

$$\partial_t \tilde{\rho}(\mathbf{k}, t) = -\lambda \int dk_1 \; \langle \tilde{G}(\mathbf{k}, t) \tilde{G}(\mathbf{k}_1, t)^* \rangle \; \tilde{\rho}(\mathbf{k}_1, t) \; . \tag{11}$$

We refer the reader to appendix A for the evaluation of $\langle \tilde{G}(\mathbf{k}, t) \tilde{G}(\mathbf{k}_1, t)^* \rangle$. The averaging necessarily introduces a temporal cut-off t_0 (which sets the implicit correlation scale of the white noise ξ). We shall only ever work to leading order in $1/t_0$. Eq.(11) now takes the form

$$\partial_t \tilde{\rho}(\mathbf{k}, t) = -\frac{\lambda D}{t_0} k^\alpha \int dk_1 \; k_1^\alpha \; \tilde{\rho}(\mathbf{k}_1, t) \; e^{-(D/2)(\mathbf{k}-\mathbf{k}_1)^2 t + i(\mathbf{k}-\mathbf{k}_1)\cdot \mathbf{R}_0} \; , \tag{12}$$

where the momentum components are indicated by a superscript for future notational convenience.

This integral equation may be recast in a more illuminating form by inverse Fourier transforming the density. After some re-arranging, one has

$$\partial_t \rho(\mathbf{r}, t) = -\frac{\lambda D}{t_0} \int dk_1 \; k_1^\alpha \; e^{-i\mathbf{k}_1 \cdot \mathbf{r}} \tilde{\rho}(\mathbf{k}_1, t) \int dk \; (k^\alpha + k_1^\alpha) e^{-(D/2)k^2 t - i\mathbf{k}\cdot(\mathbf{r}-\mathbf{R}_0)} \; . \tag{13}$$

Now the inner integral may be written as $(i\partial_{r^\alpha} + k_1^\alpha) g(\mathbf{r} - \mathbf{R}_0, t)$. The Green function $g(\mathbf{r}, t) = (2\pi D t)^{-d/2} \exp[-r^2/2Dt]$ is the probability density for the vacancy (in other words, g is the solution of the Fokker-Planck equation corresponding to Eq.(4)). One may then manipulate the outer integral over $\mathbf{k}_1$ in terms of the inverse FT of the density to obtain the following partial differential equation for $\rho(\mathbf{r}, t)$:

$$\partial_t \rho = \frac{\lambda D}{t_0} \nabla \cdot [g(\mathbf{r} - \mathbf{R}_0, t) \nabla] \; \rho \quad . \tag{14}$$

As a MFT, the above equation makes good sense. It contains the simple physical information that the OP density undergoes a diffusion process, but with the twist that the diffusivity is not constant, but proportional to the probability density of the wandering vacancy. One would have had little trouble in writing down such a MFT using physical arguments alone.

A complete analytic treatment of Eq.(14) is beyond our present remit. However, the limit $r^2 \ll Dt$ is trivially solved, since to leading order the equation reduces to

$$\epsilon(t)\partial_t \rho = \frac{D}{2}\nabla^2 \rho \ , \tag{15}$$

where $\epsilon(t) = (t_0/2\lambda)(2\pi Dt)^{d/2}$. One may solve the above equation using FT, and with the initial condition specified in Eq.(8) we obtain $\rho = A - Bg(\mathbf{r}, \tau(t))$, where $\tau(t)$ is the effective time scale of the tagged particle, and is given by $\tau(t) = \int_{t_0}^{t} dt' \ \epsilon(t')^{-1}$ which, ignoring numerical prefactors, takes the form

$$\tau(t) \sim \begin{cases} \frac{\lambda}{(Dt)^{d/2}} \frac{t}{t_0} & 0 < d < 2 \\ \frac{\lambda}{Dt_0} \ln(t/t_0) & d = 2 \\ \frac{\lambda}{(Dt_0)^{d/2}} & d > 2 \end{cases} \tag{16}$$

This solution firstly tells us that the OP density always has a gaussian envelope (at least for $r^2 \ll Dt$, which encompasses most of the physically interesting scales, since $t \gg \tau$), but that the temporal spreading of the envelope is not that of a random walker ($\Delta r \sim \sqrt{t}$), but much reduced. In two dimensions the spreading increases only logarithmically in time (as the vacancy's random walk is only just recurrent), and for $d > 2$ the spreading halts altogether after some finite time, which is accounted for by the vacancy having 'fled the scene', never to return.

The main interest in VMD is not so much in the renormalized time scale, which one can argue for on simple physical grounds, but rather in the non-gaussian nature of the OP fluctuations. These non-gaussian fluctuations were exactly calculated in lattice theories, and were generally found to have tails which decay *slower* than a gaussian. It is the aim of the following two sections to reproduce these features from the continuum theory.

IV. SOLUTION VIA PERTURBATION THEORY

We have seen the relative failure of MFT, which is essentially due to imposing a strict locality on the OP equation of motion. In fact, the evolution of the OP is non-local as seen from Eq.(5). A more systematic treatment is required to handle the subtle correlations between vacancy and OP. Given the functional nature of Eq.(5), the most useful analytic technique would appear to be perturbation theory. Since the equation of motion is linear, we shall not encounter an exponentially divergent number of terms at higher orders; rather, each order will contain only a single term.

So referring to the time integrated (and Fourier transformed) evolution equation for the OP, namely Eq.(10), we make the substitution

$$\tilde{\phi}(\mathbf{k},t) = \sum_{n=0}^{\infty} \lambda^n \tilde{\chi}_n(\mathbf{k},t) \ , \tag{17}$$

where $\tilde{\chi}_0(\mathbf{k},t) = \tilde{\phi}_0(\mathbf{k})$. Equating powers of the coupling λ yields (for $n > 0$)

$$\tilde{\chi}_n(\mathbf{k},t) = -\int_0^t dt' \ \tilde{G}(\mathbf{k},t') \int dk' \ \tilde{G}(\mathbf{k}',t')^* \tilde{\chi}_{n-1}(\mathbf{k}',t') \ . \tag{18}$$

This relation may be iterated to give the explicit solution for each order of perturbation theory as

$$\begin{aligned} \tilde{\chi}_n(\mathbf{k},t) = (-1)^n \int_0^t dt_1 \cdots \int_0^{t_{n-1}} dt_n \ \tilde{G}(\mathbf{k},t_1) \left[\prod_{m=1}^{n-1} \int dk_m \ \tilde{G}(\mathbf{k}_m,t_m)^* \tilde{G}(\mathbf{k}_m,t_{m+1}) \right] \\ \times \int dk_n \tilde{G}(\mathbf{k}_n,t_n)^* \tilde{\phi}_0(\mathbf{k}_n) \ . \end{aligned} \tag{19}$$

In principle, the solution given above may be used to calculate a range of spatio-temporal OP correlation functions. Our present aim is more modest – we shall perform a direct average of each term in the perturbation series in order to obtain the OP density $\rho(\mathbf{r},t)$. We have

$$\tilde{\rho}(\mathbf{k},t) = \tilde{\phi}_0(\mathbf{k}) + \sum_{n=1}^{\infty} \lambda^n \langle \tilde{\chi}_n(\mathbf{k},t) \rangle \ . \tag{20}$$

Using the explicit form for the $\tilde{G}$ functions as given in Eq.(7) we may write

$$\langle\tilde{\chi}_n(\mathbf{k},t)\rangle = \int\limits_0^t dt_1 \cdots \int\limits_0^{t_{n-1}} dt_n\ k^{\alpha_1} \left[\prod_{m=1}^{n-1} \int dk_m\ k_m^{\beta_m} k_m^{\alpha_{m+1}}\right] \int dk_n k_n^{\beta_n} \tilde{\phi}_0(\mathbf{k}_n)\ Q_n(\{\mathbf{k}_m, t_m; \alpha_m, \beta_m\})\ , \tag{21}$$

where the function Q_n represents the following average:

$$Q_n(\{\mathbf{k}_m, t_m; \alpha_m, \beta_m\}) = (-1)^n \left\langle \prod_{m=1}^{n} \xi^{\alpha_m} \xi^{\beta_m} \exp\left[i\mathbf{R}(t_m)\cdot(\mathbf{k}_{m-1} - \mathbf{k}_m)\right]\right\rangle\ , \tag{22}$$

with the time ordering $t_1 \geq t_2 \geq \cdots \geq t_n$, and the notation $\mathbf{k}_0 \equiv \mathbf{k}$. At this stage of the calculation we can see clearly the remaining steps. First, we must perform a multivariate average in order to determine the function Q_n. Second, we must perform the n-fold integral over the momenta $\{\mathbf{k}_m\}$. Third, we must perform the n-fold integral over the intermediate times $\{t_m\}$. Finally, we are left to resum the functions $\tilde{\chi}_n$ which will yield the FT of the mean OP density. In the absence of hindsight, it is somewhat surprising that all these steps may be performed exactly for arbitrary dimension d. The remainder of this section will consist of the details of the first two steps, whilst the third and fourth steps will be presented in the subsequent section as they are dimension-specific. Henceforth, we shall take the initial position of the vacancy to be at the origin: $\mathbf{R}_0 = \mathbf{0}$. This slight loss of generality (which is of no physical significance in the long-time regime) is more than compensated by calculational simplicity.

We begin with the first step; that of determining Q_n. We refer the reader to Appendix B in which this multivariate average is evaluated. The result is

$$\begin{aligned} Q_n(\{\mathbf{k}_m, t_m; \alpha_m, \beta_m\}) = \left(-\frac{D}{t_0}\right)^n \prod_{m=1}^{n} \Bigg\{ & \Big[\delta_{\alpha_m,\beta_m} - Dt_0(k^{\alpha_m} - k_m^{\alpha_m})(k^{\beta_m} - k_m^{\beta_m})\Big] \\ & \times \exp\Big[-\frac{D}{2}(\mathbf{k} - \mathbf{k}_m)^2(t_m - t_{m+1})\Big]\Bigg\}\ , \end{aligned} \tag{23}$$

where the symbol $t_{n+1} \equiv 0$.

To proceed with the momentum integrals, it is necessary to decide upon an initial condition. We shall use that given in Eq.(8), since our interest is presently focussed on the vacancy-driven diffusion of a tagged particle. The generic momentum integral (for $1 \leq m \leq n-1$) takes the form:

$$\int dk_m\; k_m^{\beta_m} k_m^{\alpha_{m+1}} \Big[\delta_{\alpha_m,\beta_m} - Dt_0(k^{\alpha_m} - k_m^{\alpha_m})(k^{\beta_m} - k_m^{\beta_m})\Big] \exp\Big[-\frac{D}{2}(\mathbf{k}-\mathbf{k}_m)^2(t_m - t_{m+1})\Big]$$
$$= [D(t_m - t_{m+1})]^{-1}[2\pi D(t_m - t_{m+1})]^{-d/2}\Big[\delta_{\alpha_m,\alpha_{m+1}} + D(t_m - t_{m+1})k^{\alpha_m}k^{\alpha_{m+1}}\Big]\ , \quad (24)$$

where we have worked to leading order in $(1/t_0)$. The final momentum integral involves the OP initial condition which in Fourier space takes the form $\tilde{\phi}_0(\mathbf{k}) = (2\pi)^d A\delta^d(\mathbf{k}) - B$. Its evaluation yields

$$\int dk_n\; k_n^{\beta_n}\tilde{\phi}_0(\mathbf{k}_n)\Big[\delta_{\alpha_n,\beta_n} - Dt_0(k^{\alpha_n} - k_n^{\alpha_n})(k^{\beta_n} - k_n^{\beta_n})\Big]\exp\Big[-\frac{D}{2}(\mathbf{k}-\mathbf{k}_n)^2 t_n\Big]$$
$$= \quad -Bk^{\alpha_n}(2\pi D t_n)^{-d/2}\ , \quad (25)$$

to leading order in $(1/t_0)$. Collecting our results from Eqs.(23), (24) and (25), and substituting back into (21), we have

$$\langle\tilde{\chi}_n(\mathbf{k},t)\rangle = -BD[-(2\pi D)^\gamma t_0]^{-n}\int_0^t dt_1\Bigg[\int_0^{t_1} dt_2(t_1-t_2)^{-(1+\gamma)}\cdots\int_0^{t_{n-1}} dt_n(t_{n-1}-t_n)^{-(1+\gamma)}\Bigg]t_n^{-\gamma}$$
$$\times k^{\alpha_1}\left\{\prod_{m=1}^{n-1}[\delta_{\alpha_m,\alpha_{m+1}} + D(t_m - t_{m+1})k^{\alpha_m}k^{\alpha_{m+1}}]\right\}k^{\alpha_n}\ , \quad (26)$$

where we have introduced $\gamma \equiv d/2$. An important point must be mentioned at this stage. The intermediate time integrals above appear to be divergent at one or both of their lower and upper limits. This divergence is regularized by the existence of the microscopic time scale t_0. As mentioned before, this scale appears as an effective correlation time in the white noise process. It may be taken to be arbitrarily small (with respect to any 'experimental' time scale in which one is interested). Therefore, any time integral limit is naturally softened by t_0.

We have now completed two of the four steps in the calculation of $\tilde{\rho}(\mathbf{k},t)$. To proceed further requires the evaluation of the n-fold integral over the intermediate times which is sensitive to spatial dimension, and is presented in the next section.

V. MEAN OP DENSITY IN VARIOUS DIMENSIONS

As mentioned above, we must now specify the spatial dimension of interest. In fact there are three cases: i) $d > 2$, ii) $d = 2$, and iii) $d < 2$. These cases were already apparent within MFT, and arise because of the qualitatively different behaviour of the vacancy's random walk. In case i), the vacancy will essentially 'disappear' from the vicinity of the tagged particle after a finite time. In case ii), the vacancy's walk is marginally recurrent and we expect a slow, but steady, evolution of the OP density for all times. In case iii), the vacancy's walk is 'strongly' recurrent, and thus the cross-section of vacancy-tag collisions is always 'large'. It is the aim of this final stage of the calculation to replace these qualitative descriptions by precise results. We shall analyse the three cases in turn.

A. $d > 2$

In this and the following two subsections we shall take advantage of the smallness of t_0. Integrals which are apparently divergent will be regularized using t_0, and only the most singular contribution will be retained. We stress that this form of regularization is not a mathematical manoeuvre, but is entirely consistent with the physical meaning of white noise; namely a noise process which is a limiting form of a microscopic process with a correlation time t_0.

Referring to Eq.(26) we adopt the following strategy to evaluate the integrals. First, we explicitly contract the n-fold momentum product, which will yield 2^{n-1} terms in n sets; terms in the m^{th} set being characterized by a factor of $k^{2(m+1)}$ (where m counts from 0 to $n-1$). A term in the m^{th} set will also carry a string composed of m different factors of the form $(t_j - t_{j+1})$. For each term, we perform the time integrals in order, starting from t_n, keeping only the most singular contribution at each step, and being careful to include the appropriate time-difference factors in the numerator (from the string). This procedure is fairly simple for $d > 2$, since the only integrals one encounters are

$$
\begin{aligned}
\int_{t_0}^{t-t_0} ds \, \frac{1}{(t-s)^{1+\gamma} s^{\gamma}} &\simeq \frac{1}{\gamma} \frac{1}{(t_0 t)^{\gamma}} \ , \\
\int_{t_0}^{t-t_0} ds \, \frac{1}{(t-s)^{\gamma} s^{\gamma}} &\simeq \frac{2}{(\gamma-1)} \frac{1}{t_0^{\gamma-1} t^{\gamma}} \ , \\
\int_{t_0}^{t-t_0} ds \, \frac{1}{s^{\gamma}} &\simeq \frac{1}{(\gamma-1)} \frac{1}{t_0^{\gamma-1}} \ .
\end{aligned} \tag{27}
$$

One finds that each term of a given set has the same value after integration. In detail, the m^{th} set contains C_m^{n-1} equal terms of value

$$
D^m k^{2(m+1)} \left(\frac{1}{\gamma t_0^{\gamma}} \right)^{n-m-1} \left(\frac{2}{(\gamma-1) t_0^{\gamma-1}} \right)^m \left(\frac{1}{(\gamma-1) t_0^{\gamma-1}} \right) \ .
$$

Thus, the series composed of the n sets is nothing more than a binomial series; which is trivially summed. The dominant contribution from the n-fold time integral may therefore be combined with the constant prefactor of Eq.(26) to give

$$
\langle \tilde{\chi}_n(\mathbf{k}, t) \rangle = \frac{B D k^2}{(\gamma-1)(2\pi D t_0)^{\gamma}} \left\{ -\frac{1}{\gamma t_0 (2\pi D t_0)^{\gamma}} \left[1 + \frac{2\gamma}{(\gamma-1)} D k^2 t_0 \right] \right\}^{n-1} \ . \tag{28}
$$

This ends the third step (namely the intermediate time integrals). The last step is to reconstruct $\tilde{\rho}$ from the infinite sum given in Eq.(20), using Eq.(28) above. In the present case, this sum is seen to be simple, as the series (in powers of λ) is geometric. Explicitly evaluating the sum yields the final result in the form

$$
\tilde{\rho}(\mathbf{k}, t) = A (2\pi)^d \delta^d(\mathbf{k}) - \frac{B}{2} \left[1 + \frac{1}{1 + (k/\Lambda)^2} \right] \ , \tag{29}
$$

where the momentum scale is given by

$$
\Lambda^2 = \frac{(\gamma-1)}{2 D t_0} \left[1 + \frac{(2\pi D t_0)^{\gamma} t_0}{\lambda} \right] \ . \tag{30}
$$

It is of interest to recast this result in real space. For instance, in the physically pertinent case of $d = 3$ one has

$$
\rho(\mathbf{r}, t) = A - \frac{B}{2} \left[\delta^3(\mathbf{r}) + \frac{\Lambda^2}{4\pi r} e^{-\Lambda r} \right] \ . \tag{31}
$$

Thus, for $d > 2$ we find that the initial δ-function of the OP density is evolved such that only half of its weight is smeared. That half which is smeared attains a Lorentzian profile in Fourier space, with a momentum scale Λ as given in Eq.(30). This scale is seen to be an effective UV cutoff $(1/\sqrt{Dt_0})$ renormalized by the vacancy-tag coupling λ. Note that this scale is independent of time, in accordance with our expectations. Note also that the smearing of the OP density is strongly non-gaussian. In real space, the smearing creates an OP density which is exponential in form, as seen explicitly for $d = 3$ above.

One final point: we expect that as d increases, less of the initial δ-function (in the OP density) will be smeared, since the vacancy will disappear from its vicinity with increasing efficiency. The fact that we find exactly half of the δ-function to be smeared, for all d, is a consequence of starting the vacancy's walk precisely at the location of the δ-function. Had we chosen $\mathbf{R}_0 \neq \mathbf{0}$, the d-dependence of the 'smearing fraction' would have been apparent.

B. $d = 2$

We now turn to the marginal case of $d = 2$. Within MFT we found that the root-mean-square fluctuations of the tag (i.e. the smearing of the initial δ-function in the OP density) grow as $[\ln(t)]^{1/2}$. We expect this slow growth to be retained within the exact solution. Our main interest is in how the functional form of the OP density differs from the gaussian found in MFT.

In exactly two dimensions, the n^{th}-order contribution to the OP density, as given in Eq.(26), takes the form

$$\langle \tilde{\chi}_n(\mathbf{k},t) \rangle = -BD(-2\pi D t_0)^{-n} \int_0^t dt_1 \left[\int_0^{t_1} dt_2 (t_1 - t_2)^{-2} \cdots \int_0^{t_{n-1}} dt_n (t_{n-1} - t_n)^{-2} \right] t_n^{-1} \\ \times k^{\alpha_1} \left\{ \prod_{m=1}^{n-1} [\delta_{\alpha_m,\alpha_{m+1}} + D(t_m - t_{m+1}) k^{\alpha_m} k^{\alpha_{m+1}}] \right\} k^{\alpha_n} \ . \qquad (32)$$

Our strategy for evaluating the time integrals is the same as before. We multiply out the integrand to form a total of 2^{n-1} terms arranged in n sets. Each term is integrated over the n intermediate times, with only the most singular piece retained from each integral. Care is

taken to include the appropriate factors, for a given term, in the numerator. The integrals one encounters are given below (with $p \geq 0$):

$$\begin{aligned}
\int_{t_0}^{t-t_0} ds\, \frac{[\ln(s/t_0)]^p}{(t-s)^2 s} &\simeq \frac{[\ln(t/t_0)]^p}{t_0 t} \ , \\
\int_{t_0}^{t-t_0} ds\, \frac{[\ln(s/t_0)]^p}{(t-s)s} &\simeq \frac{(p+2)}{(p+1)} \frac{[\ln(t/t_0)]^{p+1}}{t} \ , \\
\int_{t_0}^{t-t_0} ds\, \frac{[\ln(s/t_0)]^p}{s} &\simeq \frac{1}{(p+1)} [\ln(t/t_0)]^{p+1} \ .
\end{aligned} \tag{33}$$

As before, one finds that each term of a given set has the same value after integration. In this case the m^{th} set contains C_m^{n-1} equal terms of value

$$D^m k^{2(m+1)} (1/t_0)^{n-m-1} [\ln(t/t_0)]^{m+1} \ .$$

The n sets form a binomial series which is trivially summed. We find

$$\langle \tilde{\chi}_n(\mathbf{k},t) \rangle = \frac{BDk^2 \ln(t/t_0)}{2\pi D t_0} \left\{ -\frac{1}{2\pi D t_0^2} \left[1 + Dk^2 t_0 \ln(t/t_0) \right] \right\}^{n-1} \ . \tag{34}$$

The final step is to sum over the functions $\langle \tilde{\chi}_n \rangle$ as prescribed by Eq.(20). As before, this series is geometric and the sum may be immediately performed to give

$$\tilde{\rho}(\mathbf{k},t) = A(2\pi)^d \delta^d(\mathbf{k}) - B \left[\frac{1}{1 + (k/\Lambda)^2 \ln(t/t_0)} \right] \ , \tag{35}$$

where the momentum scale is given by

$$\Lambda^2 = \frac{1}{D t_0} \left[1 + \frac{2\pi D t_0^2}{\lambda} \right] \ . \tag{36}$$

Inverting the FT yields our final result

$$\rho(\mathbf{r},t) = A - \frac{B\Lambda^2}{2\pi \ln(t/t_0)} \mathrm{K}_0 \left[\frac{\Lambda r}{[\ln(t/t_0)]^{1/2}} \right] \ , \tag{37}$$

where K_0 is the modified Bessel function of zeroth order [9].

We see that the gaussian envelope for the spreading of the OP density (as found in MFT), has given way to a completely different form, namely the Bessel function K_0. This is in complete agreement with the previous exact lattice calculations [4].

C. $d < 2$

Finally we consider VMD for $d < 2$. Within lattice calculations the case of $d = 1$ may be studied either within the geometry of a chain, or within the geometry of a strip of finite width. In the former case, the situation is trivial, since the tag can only be moved back and forth to one of two sites as the vacancy passes by. In the latter, the smearing of the tag distribution function is non-trivial; the gaussian fluctuations of MFT giving way to a stretched exponential. In this subsection, we shall see how to recover these results, along with their generalization to arbitrary $d \in [0, 2]$.

Our starting point is the expression for $\tilde{\chi}_n$ given in Eq.(26). We shall adopt the same strategy as before to evaluate the n-fold integral over the intermediate times. In this case we encounter the following integrals (with $p \geq 0$)

$$\begin{aligned} \int_{t_0}^{t-t_0} ds \, \frac{1}{(t-s)^{1+\gamma} s^{(p+1)\gamma - p}} &\simeq \frac{1}{\gamma t_0^{\gamma} t^{(p+1)\gamma - p}} \, , \\ \int_{t_0}^{t-t_0} ds \, \frac{1}{(t-s)^{\gamma} s^{(p+1)\gamma - p}} &\simeq \frac{B(1-\gamma, (p+1)(1-\gamma))}{t^{(p+2)\gamma - (p+1)}} \, , \\ \int_{t_0}^{t-t_0} ds \, \frac{1}{s^{(p+1)\gamma - p}} &\simeq \frac{t^{(p+1)(1-\gamma)}}{(p+1)(1-\gamma)} \, , \end{aligned} \tag{38}$$

where $B(a, b)$ is the Beta function [9].

In the two previous subsections, we were able to extract the most singular contributions from the 2^{n-1} terms in the integrand of Eq.(26), and we found that these contributions formed a binomial series which was then easily summed. In the present case, this simple summability is lost, due to the presence of the Beta functions. However, we retain the feature that each of the C_m^{n-1} terms within the m^{th} set are equal in value. Thus, after some manipulations, we may reduce the function $\langle \tilde{\chi}_n \rangle$ to the form

$$\langle \tilde{\chi}_n(\mathbf{k}, t) \rangle = -\frac{B}{[-\gamma t_0 (2\pi D t_0)^{\gamma}]^n} \sum_{m=0}^{n-1} C_p^{n-1} \frac{[\gamma \Gamma(1-\gamma) D k^2 t^{1-\gamma} t_0^{\gamma}]^{m+1}}{\Gamma(1 + (m+1)(1-\gamma))} \, . \tag{39}$$

In order to perform the summation over m, we introduce Hankel's representation of the Gamma function [9]:

$$\frac{1}{\Gamma(z)} = \frac{i}{2\pi} \int_C d\tau \; e^{\tau} \tau^{-z} \; , \tag{40}$$

where the contour C runs from minus infinity above the negative real axis, encircles the origin clockwise, and then returns to minus infinity below the real axis. Using this representation for the Gamma function appearing in the denominator of the summand of Eq.(39), we may explicitly perform the (binomial) sum. Each function $\langle \tilde{\chi}_n \rangle$ now takes the form of an integral over τ, with n appearing only as a simple power in the integrand. Thus the sum over these functions (as dictated by Eq.(20)) is again geometric and may be performed with ease. One then has the following integral expression for the FT of the mean OP density, valid for $0 < d < 2$ (*i.e.* $0 < \gamma < 1$):

$$\tilde{\rho}(\mathbf{k}, t) = A(2\pi)^d \delta^d(\mathbf{k}) - B \frac{i}{2\pi} \int_C d\tau \; \frac{e^{\tau}}{\tau^{\gamma}} \left[\frac{1}{\tau^{1-\gamma} + (k/\Lambda)^2 (t/t_0)^{1-\gamma}} \right] \; , \tag{41}$$

where the renormalized UV cutoff is given by

$$\Lambda^2 = \frac{1}{\gamma \Gamma(1-\gamma) D t_0} \left[1 + \frac{\gamma (2\pi D t_0)^{\gamma} t_0}{\lambda} \right] \; . \tag{42}$$

We now wish to extract the scaling behaviour of the mean OP density from the above expression. For convenience we define $\delta\rho = A - \rho$, which is initially a δ-function with amplitude B. Let us first specialize to $d = 1$. In this case one may simplify the above integral considerably, using a procedure outlined in Appendix C. The result is the following scaling function:

$$\delta\rho(\mathbf{r}, t) = \frac{B}{\pi} \, \frac{z}{r} \int_0^{\infty} \frac{ds}{\sqrt{s}} \exp[-s^2 - z^2/4s] \; , \tag{43}$$

where the scaling variable is $z = r\Lambda/(4t/t_0)^{1/4}$. This integral is easily analyzed for both $z \ll 1$ and $z \gg 1$. In the former case, one finds

$$\delta\rho(\mathbf{r}, t) = \frac{B}{2\sqrt{2}\,\pi} \Lambda \left(\frac{t_0}{t} \right)^{1/4} \left[\Gamma(1/4) - 2\sqrt{\pi}\, z + O(z^2) \right] \; , \tag{44}$$

whilst in the latter, a steepest descents analysis yields

$$\delta\rho(\mathbf{r}, t) \sim \frac{B z^{2/3}}{\sqrt{\pi}\, r} \exp[-(3/4) z^{4/3}] \; . \tag{45}$$

Both of the above results are in complete agreement with the scaling functions found by Brummelhuis and Hilhorst [4], from an exact lattice calculation for VMD on an infinite strip. This gives us strong confidence in the physical integrity of our continuum theory of VMD.

For completeness we briefly describe the form of the mean OP density for arbitrary dimension $d \in [0, 2]$. The scaling variable in this case is generalized to

$$z(\gamma) = \left(\frac{1-\gamma}{2}\right)^{(1-\gamma)/2} \frac{r\Lambda}{(t/t_0)^{(1-\gamma)/2}} \ . \tag{46}$$

Referring to Eq.(41), inverse FT and subsequent analysis yields the following results. For $z(\gamma) \ll 1$ we find

$$\delta\rho(\mathbf{r}, t) = \frac{B}{(4\pi)^\gamma} \frac{\Gamma(1-\gamma)}{\Gamma(\gamma + (1-\gamma)^2)} \Lambda^{2\gamma} \left(\frac{t_0}{t}\right)^{\gamma(1-\gamma)} \left[1 + O(z^{2(1-\gamma)})\right] \ , \tag{47}$$

whilst a steepest descents analysis for $z(\gamma) \gg 1$ reveals

$$\delta\rho(\mathbf{r}, t) \sim \exp\left[-\frac{(1+\gamma)}{2} z(\gamma)^{2/(1+\gamma)}\right] \ . \tag{48}$$

This completes our study of the simplest VMD scenario; namely the effective diffusion of a tagged particle by a wandering vacancy. We have been able to give exact results for all dimensions. In the case of integer dimensions, our results are found to be in complete agreement with previous exact lattice studies.

VI. EXTENSION TO A SLAVED FLUX-LINE

In this penultimate section we shall consider a more complicated VMD scenario, both as an illustration of the utility of the coarse-grained approach, and also as a physical model for a VMD mechanism of flux lattice melting.

We consider an anisotropic lattice consisting of well separated planes (in particular, one of the high-T_c cuprates [10]). Within each plane there exists a low density of wandering vacancies. We now imagine a flux line directed perpendicular to the planes, and strongly pinned by certain lattice impurities [11]. If the binding energy is strong, thermal wandering of the line will be completely suppressed. However, a much weaker form of line wandering

may be driven by VMD of the pinning sites themselves (due to exchange with the low density planar vacancies, or vacancy aggregates). Since each plane has its own stock of vacancies, (which we disallow from hopping from plane to plane), the interactions between a given vacancy and the line are recurrent and we can expect slow and steady smearing of the mean density of the flux line. The physical existence and/or relevance of this mechanism deserves more detailed investigation (for instance, there are several competing pinning mechanisms within the material, one of which is actually due to the oxygen vacancies themselves [11]).

We shall describe this system by generalizing the continuum theory of VMD outlined in section II. First, we take for simplicity one vacancy within each plane. In the continuum, this is simply described by attaching a longitudinal coordinate to the vacancy position $\mathbf{R}$. The equation of motion for the vacancies is then given by

$$\partial_t \mathbf{R}(z,t) = \xi(z,t) \ , \tag{49}$$

where the noise has zero mean and covariance $\langle \xi^\alpha(z,t)\xi^\beta(z',t')\rangle = D\delta_{\alpha,\beta}\delta(z-z')\delta(t-t')$. The OP ϕ now describes the probability density of the flux line. It is a function of a planar coordinate $\mathbf{r}$, a longitudinal coordinate z, and time t. For a given z, the evolution of the OP is given by Eq.(5), in two dimensions. The simplest longitudinal coupling shall be taken – namely, an elastic interaction (which stems from the Josephson coupling between Cu-O planes [12]). Thus the equation of motion for the OP is

$$\partial_t \phi(\mathbf{r},z,t) = \nu \partial_z^2 \phi(\mathbf{r},z,t) - \lambda \left[\xi \cdot \frac{\partial \phi(\mathbf{R}(z,t),z,t)}{\partial \mathbf{R}(z,t)} \right] \ \partial_t \delta^d(\mathbf{r} - \mathbf{R}(z,t)) \ , \tag{50}$$

where ν is an effective longitudinal elasticity.

As an initial condition, the simplest choice is to take the flux line to be straight, and located at the origin: $\phi(\mathbf{r},z,0) = A\delta(\mathbf{r})$. Also, we could (artificially) start all the vacancies at the origin: $\mathbf{R}(z,0) = \mathbf{0}$.

This model may now be analysed in precisely the same way as our original VMD model; namely through an infinite order perturbation expansion in powers of λ. One must use an additional longitudinal FT to diagonalize the elastic coupling. The appearance of multiple longitudinal Green functions makes the explicit evaluation of the functions $\langle \tilde{\chi}_n \rangle$ more

challenging than before. However, analytic progress seems possible and is currently being pursued. It is certainly of interest to calculate the ν-dependence of the mean OP evolution, to see how effectively the flux line elasticity combats the driving forces of VMD. Preliminary results indicate that i) for $\nu \to \infty$ the line fluctuations become gaussian and coincide exactly with the fluctuations of a single point within MFT (as described in section III), and ii) for $\nu \to 0$ the line fluctuations have a *singular* dependence on the elasticity (and thus differ from the fluctuations of independent planar tags as described in sections IV and V).

VII. CONCLUSIONS

In this paper we have constructed and solved a continuum theory of vacancy-mediated diffusion. Our main intention has been to test the theory against exact results known from lattice studies [4,5]. In particular we have thoroughly examined the evolution of the mean OP density in the case of a single vacancy smearing an originally sharply-peaked OP fluctuation (which corresponds to the lattice scenario of following the motion of a tagged particle due to vacancy exchange). At the level of mean field theory, we found the (almost obvious) length-time scaling for the OP density, but no sign of the non-gaussian fluctuations, which were the most interesting results obtained from the lattice studies. Therefore we pursued a more systematic treatment, based on an infinite order perturbation expansion. We presented an exact analysis of our theory in all dimensions, and complete agreement has been found with the dynamical scaling results obtained from the lattice. In particular, we find that for $d > 2$, the OP density is smeared over a limited range and then freezes. The envelope is a simple exponential. In $d = 2$, the smearing is slow, but continues indefinitely. The envelope is described by the modified Bessel function K_0. Finally, for $d < 2$, a more challenging calculation revealed that the envelope of OP smearing is described by a stretched exponential. Our results have the advantage of being valid for arbitrary dimension, thus revealing more clearly their analytic structure. In the final section we proposed an extension of simple VMD to the diffusion of a pinned flux line, slaved to planar vacancy exchange.

Analysis of this more challenging problem is in progress.

There are, needless to say, many other extensions to the current work. We mention the more obvious here. First, it would be interesting to calculate higher order OP correlations. Throughout the present work, we have concentrated on the mean OP density. However, there is much non-trivial information hidden in the simplest spatio-temporal correlation functions. Second, one can apply the continuum theory to more complicated (single vacancy) scenarios; mainly by adjusting the boundary and initial conditions. For instance, a step function initial condition would correspond to the vacancy-mediated roughening of an initially straight domain wall. Finally, one could investigate simple mechanisms whereby the random walk of the vacancy itself is weakly coupled to the OP distribution, which is a physically relevant perturbation.

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APPENDIX A:

In this appendix we explicitly evaluate the average $\langle \tilde{G}(\mathbf{k},t)\tilde{G}(\mathbf{k}_1,t)^*\rangle$, where G is defined in Eq.(7). Aside from momentum prefactors, we need to evaluate

$$I_{\alpha,\beta}(\kappa,t) = \left\langle \xi^\alpha(t)\xi^\beta(t)\exp\Big[i\int_0^t dt'\ \xi(t')\cdot\kappa(t')\Big]\right\rangle . \tag{A1}$$

We have generalized the momentum in the exponent to a time-dependent form $\kappa(t)$ for a reason soon to become clear. At the end of the averaging procedure we shall reset $\kappa = \mathbf{k}-\mathbf{k}_1$ as required.

The average given above is most easily evaluated by generating the noise prefactors via functional differentiation with respect to $\kappa(t)$. Thus,

$$\begin{aligned}
I_{\alpha,\beta}(\kappa,t) &= -\frac{\delta^2}{\delta\kappa^\alpha(t)\delta\kappa^\beta(t)}\left\langle \exp\Big[i\int_0^t dt'\ \xi(t')\cdot\kappa(t')\Big]\right\rangle \\
&= -\frac{\delta^2}{\delta\kappa^\alpha(t)\delta\kappa^\beta(t)}\exp\Big[-(D/2)\int_0^t dt'\ \kappa(t')^2\Big] \\
&= \frac{D}{t_0}\Big[\delta_{\alpha,\beta} - Dt_0\kappa^\alpha(t)\kappa^\beta(t)\Big]\exp\Big[-(D/2)\int_0^t dt'\ \kappa(t')^2\Big] ,
\end{aligned} \tag{A2}$$

where t_0 is the implicit scale of the noise correlations. We need only retain the first term, given the smallness of t_0. Using this result with Eq.(11) yields Eq.(12) in the main text.

APPENDIX B:

In this appendix we outline the evaluation of Q_n as defined in Eq.(22). The terms in the exponent of Q_n are of the form $\mathbf{R}(t_m)\cdot(\mathbf{k}_{m-1}-\mathbf{k}_m) = \int_0^{t_m} ds_m\ \xi(s_m)\cdot(\mathbf{k}_{m-1}-\mathbf{k}_m)$. We replace the momenta appearing in the exponent with generalized momenta $\kappa_m(s_m)$. After the averaging procedure we set $\kappa_m(s_m) = \mathbf{k}_{m-1}-\mathbf{k}_m$. This allows us to generate the noise prefactors from functional differentiation. Thus we have

$$Q_n(\{\kappa_m,t_m;\alpha_m,\beta_m\}) = \left[\prod_{m=1}^n \frac{\delta^2}{\delta\kappa_m^{\alpha_m}(t_m)\delta\kappa_m^{\beta_m}(t_m)}\right]\ \left\langle \exp\left[i\sum_{m=1}^n\int_0^{t_m} ds_m\ \xi(s_m)\cdot\kappa_m(s_m)\right]\right\rangle . \tag{B1}$$

The average appearing above is easily performed over the multivariate gaussian noise distribution, and yields

$$\left\langle \exp\left[i\sum_{m=1}^{n}\int_{0}^{t_m} ds_m\ \xi(s_m)\cdot\kappa_m(s_m)\right]\right\rangle = \exp\left\{-\frac{D}{2}\sum_{m=1}^{n}\int_{t_{m+1}}^{t_m} ds\ \left[\sum_{l=1}^{m}\kappa_l(s)\right]^2\right\} \ . \tag{B2}$$

A given double functional derivative of the exponent gives:

$$\begin{aligned}
&\frac{\delta^2}{\delta\kappa_m^{\alpha_m}(t_m)\delta\kappa_m^{\beta_m}(t_m)}\exp\Big\{-(D/2)\int_{t_{m+1}}^{t_m} ds\ [\mathbf{K}(s)+\kappa_m(s)]^2\Big\} \\
&\qquad = -\frac{D}{t_0}\Big[\delta_{\alpha_m,\beta_m} - Dt_0(K^{\alpha_m}(t_m)+\kappa^{\alpha_m}(t_m))(K^{\beta_m}(t_m)+\kappa^{\beta_m}(t_m))\Big] \\
&\qquad\qquad \times\exp\Big\{-(D/2)\int_{t_{m+1}}^{t_m} ds\ [\mathbf{K}(s)+\kappa_m(s)]^2\Big\} \ .
\end{aligned} \tag{B3}$$

We use this last result to perform the n double functional derivatives in Eq.(B1). We note that using our final replacement for $\{\kappa_m\}$ yields $\sum_{l=1}^{m}\kappa_l = \mathbf{k}-\mathbf{k}_m$. Thus we reproduce Eq.(23) as given in the main text.

APPENDIX C:

In this appendix we give a brief analysis of the integral appearing in Eq.(41), specializing to $d = 1$. In terms of the density difference, this has the form

$$\delta\tilde{\rho}(\mathbf{k},t) = B\frac{i}{2\pi}\int_C d\tau\ \frac{e^\tau}{\tau^{1/2}}\left[\frac{1}{\tau^{1/2}+b(k,t)}\right] \ , \tag{C1}$$

where for convenience we have set $b = (k/\Lambda)^2(t/t_0)^{1/2}$. First, we change variables from τ to $-x$ (which runs along the negative real axis) by integrating across the branch cut. This yields

$$\delta\tilde{\rho}(\mathbf{k},t) = \frac{B}{\pi}\int_0^\infty \frac{dx}{\sqrt{x}}\ \frac{e^{-b^2x}}{(1+x)} \ . \tag{C2}$$

Next we change variables to $y = \sqrt{x}$. Rewriting the exponential using a Hubbard-Stratonovich transformation gives the double integral

$$\delta\tilde{\rho}(\mathbf{k},t) = \frac{B}{\pi}\int_{-\infty}^{\infty}\frac{dy}{(1+y^2)}\ \int_{-\infty}^{\infty}\frac{ds}{\sqrt{\pi}}\ e^{-s^2+2ibsy} \ . \tag{C3}$$

The integral over y now resembles a Fourier transform, and results in a simple exponential, so that

$$\delta\tilde{\rho}(\mathbf{k},t) = B \int_{-\infty}^{\infty} \frac{ds}{\sqrt{\pi}}\ e^{-s^2-2b|s|}\ . \tag{C4}$$

The inverse Fourier transform from k to r is now easily performed resulting in Eq.(43), as shown in the main text.

For general $d \in [0,2]$, the analysis is more difficult. Progress is made by exponentiating the Lorentzian form in Eq.(41) using an auxiliary integral, and then performing the asymptotic expansions on the resulting double integral.

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